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(54) Title: WATER GAS SHIFT REACTOR AND HEAT EXCHANGER (57) Abstract An innovative water gas shift reactor is disclosed in which the shift reactor is maintained at near constant temperatures and/or is continuously humidified in manners that attain low exit temperatures at elevated humidity. These conditions achieve minimum carbon monoxide concentrations in the exit gas. This accomplished either isothermally with constant water vapor pressure or over a range of controlled temperatures and controlled water vapor pressures. Several alternate hardware configurations and an overall method to minimize carbon monoxide in the product gas are taught. This innovative shift reactor is related to underoxidized burner (UOB TM), partial oxidation reformers, shift reactor and fuel-cell assemblies.		

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WATER GAS SHIFT REACTOR AND HEAT EXCHANGERField and Background of the Invention

This invention relates to water gas shift reactors and/or
5 heat exchangers of the type that reduce or minimize the
concentration of carbon monoxide and/or increase the level of
hydrogen in gas streams. Such gas streams may be produced by the
mixing of air and fuel and the burning thereof in underoxidized
burners, partial oxidation reformers, fuel-cell systems, and the
10 like. The present invention specifically addresses water gas
shift reactors of the type having constant temperature heat
transfer devices, or heat/mass exchangers, therein. The
invention may be used in combination with a hydrocarbon reformer
system, such as an underoxidized burner assembly, as well as
15 fuel cell assembly, such as a polymer electrolyte membrane
(PEM).

The temperature reduction of a gas stream being treated, as
well as the increased humidity of such gas stream, are also both
aspects of the present invention. The heat exchanger or shift
20 reactor of the present invention receives and continuously
humidifies a process gas in a manner that can achieve lowered
temperatures of the process gas as well as elevated humidity
levels thereof.

Underoxidized burners (UOB™) described in several of
25 Applicant's previous U.S. patents including 5,207,185;
5,299,536; 5,441,546; 5,437,123; 5,529,484; and 5,546,701 (all
of which are incorporated herein by reference) disclose
apparatus and methods for producing hydrogen for emissions
control in an internal combustion process, and for fuel cells
30 used in power generation systems. Such apparatus may have
associated therewith a downstream catalytic shift reactor that
converts carbon monoxide (CO) formed in the underoxidized
burner, in the presence of water vapor, to hydrogen and carbon
dioxide. The catalytic shift reactor serves a dual advantageous
35 role, namely, increased hydrogen concentrations and decreased
carbon monoxide concentrations.

In conventional processes, the product gas produced from a

reformer or burner is quenched with water, thereby increasing its humidity and decreasing its temperature prior to entering an adiabatic shift reactor. The chemical reaction in the shift reactor releases thermal energy and consumes water. This causes
 5 the water vapor to decrease and the temperature of the process gas to increase, both of which raise the equilibrium CO concentration to higher levels.

The theoretical thermal output of the shift reaction, which is represented as $\text{CO} + \text{H}_2\text{O} = \text{H}_2 + \text{CO}_2 + \text{heat}$, is about 9 Kcal per
 10 g-mole of hydrogen, which normally causes an increase of about $50^\circ\text{--}100^\circ\text{C}$ ($100^\circ\text{--}200^\circ\text{F}$) in shift catalyst temperature. At equilibrium, the extent of the shift reaction decreases as the temperature rises, and increases with water vapor content. This is illustrated in Table 1 below. The Table shows the amount of
 15 water added to the product gas, the equilibrium temperature after the shift reaction and sensible cooling, the net hydrogen production, and the equilibrium concentration of CO in the product gas which exits from the system.

20 Table 1. Theoretical Gas Equilibrium CO Concentrations
 (Methane + .45 Stoichiometric Air + Liquid Water, Feeds @ 78°F)

Water Added	Temperature	Product Moles	CO concentratn
<u>$\text{H}_2\text{O}(l)/\text{CH}_4$</u>	<u>$^\circ\text{F}$</u>	<u>H_2/CH_4</u>	<u>ppm CO</u>
5.8	193	2.20	6.1
25 5.5	214	2.20	9.7
5.0	328	2.20	74
4.0	580	2.19	1,417
3.0	864	2.10	10,099
2.0	1184	1.90	35,261
30 1.0	1577	1.63	77,178
0.0	2112	1.37	130,220

The temperatures indicated in Table 1 are those attained after thermal input from the shift reaction (see the shift
 35 reaction above, which produces heat). In the present invention, the exchanger apparatus, as will be described more fully below, allows for the movement and use of this heat produced by the

shift reaction in the evaporation process. This, in turn, facilitates attainment of low CO concentrations.

As mentioned above, the addition of water has the effect of increasing the amount of hydrogen produced, at the same time decreasing the quantities of carbon monoxide. As shown in Table 1, near maximum hydrogen production is achieved when water in the range of 4 moles/mole of methane is added, which results in equilibrium temperatures of about 500°F. In order to achieve minimum CO concentrations, a maximum amount of water is needed. In the Table 1 above, this is about 5.8 moles of water per mole of methane. The equilibrium temperature is about 193°F, and the product gas exits the system near saturation. In this way, the maximum practical water addition is achieved.

15 Summary of the Invention

The present invention is for a shift reactor and exchanger, as well as methods therefor, in which a process gas is continuously humidified in a manner that achieves low exit temperatures from the system of the process gas, at elevated humidity levels. These conditions are favorable since they achieve minimum carbon monoxide concentrations and/or maximum hydrogen levels in the gas exiting the system. These conditions are achieved isothermally with constant water vapor pressure, or over a range of controlled temperatures with controlled water vapor pressures. Various structural embodiments and different methods are disclosed in this invention to minimize carbon monoxide levels in the exiting gas from the system.

Isothermal conditions are achieved by providing a constant temperature heat exchanger device in association with a shift reactor, the heat exchanger device comprising sealed passages containing only a liquid, such as water and its vapors, and means, such as mechanical means, to circulate the enclosed liquid vapor mixture. The device may supply thermal energy to a water reservoir in order to continuously evaporate water, and thereby provide a constant vapor pressure that passes through the shift reactor. The apparatus may add or remove thermal energy from the shift. Removing thermal energy decreases CO

content. The constant temperature apparatus can also be integrated with a downstream fuel cell to manage its temperature and provide added thermal energy to the water evaporation process.

5 In another embodiment, structures are provided for controlling temperature and water pressure using a "heat/mass" exchanger integrated into the shift reactor. A heat/mass exchanger permits the exchange of both heat produced by chemical reactions, and mass, such as water vapor. As will be described
10 in further detail below, such apparatus comprises ducts, tubes or barriers whose walls are composed of porous, membranes or materials. In this embodiment, a process gas stream, consisting of mixed gases containing carbon monoxide, hydrogen and other gases, flows on one side of the membrane, which may, for
15 example, be a hydrophobic membrane. A liquid, which serves as a coolant stream and generally comprises water, flows on the opposite side of the hydrophobic (or other) membrane. The hydrophobic (or other) membrane is selected such that liquid is prevented from passing through the membrane, and at the same
20 time facilitates the transfer of thermal energy from the process gas stream into the water stream. This transfer of thermal energy causes the evaporation of water to water vapor, which is permitted to freely flow through the hydrophobic membrane and humidify the process gas, which flows through a catalyst on the
25 other side of the hydrophobic membrane. In a preferred embodiment, the coolant stream of liquid and the process gas stream flow in counter-current direction with respect to each other. The evaporation process cools the shift reactor, and the added water vapor increases the humidity level or water vapor
30 partial pressure within the process gas stream.

Reference is now made to Figure 1 of the drawings, which illustrates various equilibrium CO concentrations as a function of the product gas temperature and water vapor pressure (humidity). The various equilibrium CO concentrations have been
35 plotted based on an underoxidized burner reformer operating on gasoline and air with a stoichiometric ratio of 0.37 and a pressure of 16 psia. The figure shows a series of curves each

representing equilibrium CO concentrations from 15 ppm - 500 ppm. The dark solid line in the graph represents the water saturation partial pressure as a function of temperature.

In Figure 2 of the drawings, the same data as shown in Figure 1 is illustrated once more, but Figure 2 also illustrates low temperature shift (LTS) processes of various types. In the conventional LTS process, the product gas is cooled and then passes through an adiabatic reactor in which the temperature increases and the humidity level decreases as a result of the shift reaction ($\text{CO} + \text{H}_2\text{O} = \text{H}_2 + \text{CO}_2 + \text{heat}$). Figure 2 also shows an improved cooled LTS process, in which the reactor process gas is cooled as the process gas moves towards the exit. Lower CO levels are achievable with this cooled LTS process.

Figure 2 also shows the humidified LTS (HLTS) process of the invention. In this process, the humidity level, or water vapor pressure, continually increases as a result of the effects of the isothermal heat exchanger or the heat/mass exchanger of the present invention. The method of this innovative process comprises the duality of the cooling and humidifying of the process gas as a parallel process, as it progresses towards the exit.

It is well known that the inefficiency of a fuel cell results in significant heat liberation. For example, a 50 kilowatt, 50% efficient fuel cell liberates 40,000 kcal per hour (170,000 btu/hour) of thermal energy, and this can cause a considerable temperature rise. High temperatures can harm cell components, and large temperature gradients within the system can cause poor utilization of the active fuel cell areas. For these reasons, fuel cells are normally cooled to a controlled temperature at the most satisfactory reaction kinetics. In PEM fuel cells, this is typically achieved by water which is circulated through internal passageways. The effectiveness of this cooling mechanism varies according to the flow rates and uniformity of the water flow, and, in practice, constant temperature over the fuel cell is rarely achieved. While better temperature constancy can be accomplished with heat pipes, these depend on capillary effects to circulate their internal media.

This often results in insufficient flow rates. In either type of cooling, whether the heat exchange or heat pipe, major temperature control is normally achieved via a coupled external heat exchanger that dissipates the removed thermal energy to the environment. Such a system requires critical design attention.

It is an aspect of the present invention, that the heat transfer devices disclosed, when integrated with both the LTS reactor and a fuel cell, provide simpler and better controlled means to achieve cooling, and maintain optimum temperatures. If the fuel cell heat can be effectively converted to humidity, the fuel processing heat can be used to superheat the water to vapor. In such a case, the water vapor partial pressure, or humidity levels, within the LTS reactor could easily be doubled from levels illustrated in Figure 2. Therefore, equilibrium CO concentrations of under 15 ppm can be achieved.

Brief Description of the Drawings

Figure 1 is a graph showing equilibrium CO concentrations as a function of the product gas temperature and water vapor pressure;

Figure 2 shows equilibrium carbon monoxide concentrations, and comparison of various low temperature shift processes, once more as a function of the product gas temperature and the water vapor pressure;

Figure 3 is a cross-sectional schematic view through an isothermal heat exchanger in accordance with the present invention;

Figure 4 is a cross-section through an isothermal heat exchanger and shift reactor of the present invention;

Figure 5 is a cross-section showing a detail of a shear mixing and recirculating means in accordance with the invention;

Figure 6 is another embodiment of a shift reactor for use with a heat exchanger, in accordance with the present invention;

Figure 7 is a diagrammatic cross-section through a heat/mass exchanger, according to one embodiment of the invention;

Figure 8 is a cross-section through a plurality of

heat/mass exchangers of the type shown in Figure 7;

Figure 9 is a cross-section through a heat/mass exchanger in accordance with a further embodiment of the invention;

Figure 10 is a diagrammatic representation showing a heat/mass exchanger and the reactions which take place therein;

Figure 11 is a cross-section through a heat/mass exchanger according to one embodiment of the invention, including multiple assembly sections, as well as water and process stream supply and removal.

Detailed Description of the Preferred Embodiments

Two examples showing embodiments of the present invention comprise the constant temperature device on the one hand, and the heat/mass exchanger (including a selectively porous membrane, such as a hydrophobic membrane) on the other.

1. Constant Temperature Device

In one aspect, the invention teaches an isothermal heat exchanger which is in thermal contact with a shift reactor and/or fuel cell. The isothermal heat exchanger may consist of a thin walled, large surface area container filled with a liquid, such as water, and its vapors, which are mechanically circulated. When thermal energy is added or removed, respectively, from any one portion or section of the exchanger, that increment of liquid-vapor immediately adjacent the section where thermal energy is added or removed undergoes spontaneous vaporization (energy added) or condensation (energy removed). The increments are circulated and move about within the exchanger, and as they move, they subtract thermal energy at hot areas in the exchanger by boiling and thereby absorbing the thermal energy, or add thermal energy at cool areas by condensing and giving off such thermal energy in these cool areas. Buoyancy will induce minor, slow circulation, which varies with elevation and has horizontal movement. In a preferred version, the isothermal heat exchanger of the invention incorporates a pump that induces or provides continuous circulation of the liquid-vapor media within the system.

This arrangement results in virtually constant temperature over the surface of the area container, despite very large inputs and/or withdrawals of energy which may take place at the surface. Moreover, the operation of the device is not affected by the height of the points where heating or cooling takes place.

As will be described further below with reference to the drawings, the device somewhat resembles a heat pipe, but its internal process does not depend upon capillary effects, which have the unfavorable consequence of "starving". Moreover, in maintaining a set temperature, heating or cooling coils may be located within the exchanger, in direct contact with the enclosed liquid-vapor media. Such coils improve the capacity to control the isothermal heat exchanger, in contrast with conventional means that use external heat exchange surfaces, thereby separating the unit from its controls.

2. Heat/Mass Exchanger (hydrophobic membrane)

In another form, a structure and method is provided for cooling the low temperature shift and simultaneously adding water. This is achieved by circulating water through a heat/mass exchanger that is embedded in, for example, a shift reactor. As will be described in further detail below with reference to the drawings, the water preferably flows in a counter-current direction to a process gas. Surfaces in the heat/mass exchanger between the water and process gas flow respectively are composed of certain porous materials, for example, a hydrophobic layer. Such materials constitute a barrier to liquid water, but are porous to water vapor. Such materials may include, for example, Gortex™ and carbon graphite or porous nickel substrates treated with Teflon™ or other hydrophobic materials. In this device, the circulating water is heated by absorbing thermal energy generated by the shift reaction in the process gas. This thermal energy causes evaporation of water that in turn humidifies and cools the shift reactor. The vapors pass through the hydrophobic materials or membrane, since such membrane is porous to water vapor, and into the flowing process gas thereby providing increased humidity for the shift reaction. Various forms of the

heat/mass exchanger may be provided, including one which is of a spiral configuration, or others which may constitute a plurality of horizontal sections with openings on the side connected to inlet and outlet manifolds through which the water or process gas, respectively, can flow.

As mentioned, a preferred embodiment of the invention includes a heat/mass exchanger where the water flow is counter-current to the process gas flow through a catalyst bed. Water enters at the top of the exchanger at its lowest temperature (for example, 180°F). A low temperature is, of course, preferred since it results in minimum CO production and maximum hydrogen production. The partially hydrated process gases enter the system at temperatures which are low enough, but also acceptable to the catalyst, to thereby induce the shift reaction. As the water moves down the heat exchanger, it absorbs heat from the shift reaction to raise the water temperature and therefore promote evaporation. The water flow finally exiting at the bottom or opposite end of the reactor from which it enters is at a higher temperature than the water entering the reactor. The shift process gases move upward, receiving further vapor from the evaporated water as they do so, and the shifted product emerges at the top of the reactor with higher humidity, lower temperatures and minimum CO concentrations.

With reference to Figure 3 of the drawings, there is shown a schematic section through a constant temperature heat exchanger of the invention. In this Figure, although only certain tubes and passages are shown, several may be used so as to provide a birdcage type structure.

The constant temperature heat exchanger, generally indicated by reference numeral 10, comprises a thin walled, large surface area housing container 12 defining a chamber 14. Within the chamber 14, there is located a upper circular tube 16, located near an upper portion of the exchanger, and a lower circular tube 18, located near the lower end of the chamber 14. The tube 16 defines a passage 20 running therethrough, while the tube 18 defines passage 22.

The upper and lower circular tubes 16 and 18 are connected

to each other by one or more, and preferably a plurality, of vertical tubes 24, two of which are shown in Figure 3 of the drawings. The vertical tubes 24 may be linear, spiraled, or have some other shape suitable which may be selected. Each vertical tube 24 defines a passage 26, and the passages 26 are in communication with the passages 20 and 22 so that a liquid vapor-medium therein, as will be described further below, can circulate freely around and between the upper and lower circular tubes 16 and 18, and the vertical tubes 24.

Within the passage 20, there is located an inner circular tube 28, the inner circular tube 28 defining a passage 30. Within the passage 22, there is located an inner circular tube 32, which defines a passage 34. The inner circular tubes 28 and 32 respectively are joined to each other by a side arm tube 36, the side arm tube 36 having a passage 38 which is in communication with the passages 30 and 34, and which permits a water-vapor medium therein, as will be discussed further below, to circulate freely through the passages 30, 34 and 38. As pump 40 is located in the side arm tube 36 to facilitate circulation of the water-vapor medium through these passages.

The vertical tubes 24, at their ends, are accommodated in holes 42 drilled in the circular tubes 16 and 18 respectively, and appropriate sealing between the vertical tubes 24 and the circular 16 and 18 are provided. The side arm tube 36 has upper and lower ends 44 and 46 respectively which pass through holes 48 in the circular tubes 26 and 18 respectively, where appropriate sealing takes place, the very ends 44 and 46 of the side arm tube 36 fitting into holes drilled into inner circular tube 28 and 32, to permit circulation of the water-vapor medium, as mentioned above.

The passages 30 and 34 in the inner circular tubes 28 and 32, as well as the passage 26 in the vertical tube 24 are all filled with a water-vapor medium, providing a closed circuit in which the water-vapor medium circulates. This circulation is facilitated or provided by the pump 40.

A control tube 50 is also located within the passage 20 of the upper circular tube 16. The control tube 50 is thus circular

in structure, and has two ends 52 and 54 which extend outwardly from the circular tube 16. The ends 52 and 54 are connected to an external source so that the water-vapor medium, which is provided within the passage 56 of the control tube 50, can
5 circulate through this external source, which may provide cooling or heating effects to facilitate control of the temperature within the heat exchanger.

The passage 20 and passage 22 in the upper and lower circular tubes 16 and 18 respectively are also filled with a
10 water-vapor medium, the medium extending through the passages 26 of the vertical tubes 24 as well. The water-vapor medium fills these passages such that the level of the water is above the control tube 50. A valve 58 located on an exit tube 60 provides an outlet for all gaseous components, other than vapor from the
15 liquid-vapor medium circulating in the various passages.

In one particular embodiment of a heat exchanger of the type shown in Figure 3, the upper and lower circular tubes 16 and 18 comprise a pair of tubes approximately 12 inches long and approximately 1 inch in diameter. The vertical tubes 24 each
20 comprise a tube approximately 18 inches long and about 0.5 inches in diameter, joined at their ends 42 in suitable manner to the circular tubes 16 and 18. The inner circular tubes 28 and 32 comprises tubes about 12 inches long and approximately 0.25 inches in diameter, and these are, of course, located within the
25 circular tubes 16 and 18 respectively. The holes drilled in all of these tubes to effect appropriate connections in the manner described above are sealed at their ends to ensure that the water-vapor medium does not leak. In filling the unit and the various tubes with water, extraneous gases are evacuated, at
30 least in part through the valve 58 in the exit tube 60, which leads to the outside environment.

In an embodiment, the pump 40 circulates the internal water-vapor medium in the passages 38, and passages 30 and 34, at about 50 ml/min. To test the system, heated air at
35 approximately 1,400°F from a 0.5 kW electric torch was directed at a 1 inch section of the circular tube 16. Thereafter, temperatures were measured at various points in the surface of

the heat exchanger. As heating continued, all temperatures were found to remain within about 1°F of each other. Final equilibration within the room temperature environment was at 190±1°F at all points on its surface. In another test, while the electric torch heated the same small section of circular tube 16, the device was cooled by cold tap water, passing over 2 inches of section of the circular tube 18. Measurements once more showed constant overall temperature range of ±1°F during the cooling process. Final equilibration was measured at 95±1°F. The heating and cooling areas were then reversed, with the electric torch directed on to the lower circular tube 18, and tap water passed over the upper circular tube 16. Measured temperatures again were constant throughout until equilibration occurred. The results outlined above confirm that the heat exchanger preserves temperature constancy despite thermal inputs or decrements at any point on its surface.

Reference is now made to Figure 4 of the drawings which shows schematically a heat exchanger of the type shown in Figure 3 incorporated into a shift reactor and located above and upstream of an underoxidized burner, designated by the references numeral 62. With respect to the components of the heat exchanger already described in Figure 3, the same reference numerals will be given to like components in Figures 3 and 4 respectively.

In Figure 4, the heat exchanger includes an insulation layer 64 located between the underoxidized burner 62 and a shift reactor, as will be described. The insulation layer 64 is fitted against a metal plate 66 that separates the underoxidized burner from the shift reactor. Catalyst pellets 68 are placed in the chamber 14, between the vertical tubes 24, with the catalyst pellets 68 being confined within the heat exchanger by an upper screen 70 at the top of the heat exchanger and a lower screen 72 at the bottom of the heat exchanger. Effluent from the underoxidized burner passes through outlet 74 into a tube 76 shown in both Figures 4 and 5 of the drawings. Figure 5 shows the tube 76 as well as, in cross-section, those structures located within the tube 76. The tube 76 is closed at its top end

78.

A spray outlet 80 directs a spray of liquid water into the hot effluent from the underoxidized burner. The effluent with vaporized water leaves the tube 76 through nozzles 82, which are downwardly directed into annulus 84 defined by the space between tubes 76 and 86. It will be noted that the top end 88 of tube 86 is above the upper catalyst screen 70, and the lower end 90 is below the lower screen 72. The heat exchanger may be filled with water up to a level generally indicated by reference numeral 92 although this is optional. If water is added, the water would fill the space above the metal plate 66, with the upper surface water level 92 covering, and being above, the circular tube 18 of the heat exchanger. The open lower end 90 of the tube 86 is below the water level 92, and above the circular tube 18.

In operation, hot gas products from the underoxidized burner enter through outlet 74 and move into the tube 76, where they meet a water spray emerging from the spray outlet 80. The mixture of vaporized water and underoxidized burner effluent leave the tube 76 through nozzles 82, and enter the annulus 84 where they educt a portion of the shift products from the top of the catalyst bed, thus providing, at least in part, recirculation of the gas stream. In the embodiment where water is filled in the bottom of the heat exchanger to the water level 92, the mixture from the annulus 84 will bubble under the surface of the water, where thermal equilibration takes place. Then, the non-condensable gases and water vapor at the vapor pressure related to the temperature will move upward through the lower screen 72 and through the catalyst pellets 68. As water is moved by the shift reaction, it is automatically replaced by water vaporized from the bottom, whose temperature is maintained constant, as discussed later, with the resultant constant vapor pressure.

If water is not filled so as to cover the circular tube 18, the mixture from the annulus 84 will pass upward through the catalyst pellets 68. The passage of the gas and water vapor mixture through the catalyst results in the shift reaction, as follows: $\text{CO} + \text{H}_2\text{O} = \text{H}_2 + \text{CO}_2 + \text{heat}$. Through normal heat

exchange methods, the thermal energy liberated by the reaction is rapidly absorbed by the vertical tubes 24. It is then transferred through the shift reactor to maintain constant temperature. If the space is filled with water to water level 5 92, the thermal heat from the shift reactor will also be returned thereto, to provide further thermal energy to effect vaporization at constant temperature.

This process of heat transfer maintains the catalyst pellets 68 and water reservoir at constant temperature, and 10 supplies part of the heat required to vaporize the water. If necessary, heating or cooling the water in the water-vapor medium, in order to maintain a desired temperature, can be achieved by treating the water-vapor medium in the control tube 50 by circulating this water-vapor to an external source through 15 ends 52 and 54 respectively, and heating or cooling the water therein so that its circulation within the circular tube 16 will have the effect of bringing the water-vapor medium in contact with the control tube 50 to a desired temperature level, and thereafter circulating this level into the remainder of the 20 system.

The shift products, particularly the hydrogen whose content has been increased, as well as carbon dioxide, leave through an opening 94, as illustrated in Figure 4.

As is now described with reference to Figure 5 of the 25 drawings, carbon formation can be eliminated or much reduced by adding liquid water to the underoxidized burner effluent. When liquid water is added to the hot effluent to produce steam for the shift reactor, the effluent is cooled as water vaporizes. In conventional systems, when water spray is introduced as a spray, 30 the cooling process is slow. The effluent-water mixture may be at intermediate temperatures for relatively long time periods, and, at these intermediate temperatures, the CO disproportionation reaction is known to occur. The disproportionation reaction is represented as follows: $\text{CO} = 0.5$ 35 $\text{CO}_2 + 0.5\text{C}$. In other words, the intermediate temperatures result in the disproportionation of carbon monoxide into carbon dioxide and the production of solid carbon. Solid carbon production in

the system is undesirable since it attaches to the system, causing build-up and related problems.

One aspect of the invention, as particularly illustrated in Figure 5, uses a shearing process instead of a spray, thereby accelerating the cooling of the effluent, and greatly minimizing the production of solid carbon which may result from the disproportionation process. Figure 5 shows an embodiment of shear mixing and recirculation hardware. In Figure 5, the outlet 74 conveys the UOB™ effluent from the burner. A disc 96, whose diameter is greater than the internal diameter of outlet tube 74, is located above this outlet 74. A tube 98, whose outer diameter is smaller than that of the disc 96, is sealed at the bottom by a sealing disc 100, which has the same outer diameter as, and is located just above and close to, the disc 96. A water entry tube 102 passes through the inside of the tube 98, and is shielded by this tube 98. The exit of the water entry tube 102 is attached to the sealing disc 100, and water is able to emerge from the bottom of the water entry tube through the hole 104 bored in the middle of the sealing disc 100. A gap 106 between the discs 96 and 100 is provided, and this gap may be empty, or filled with sponge-like porous material such as certain ceramic insulation, metal screen or the like.

A tube 108, of larger diameter than tube 98, is fitted around the tube 98. The bottom of the tube 108 is attached to the metal plate 66, while its top is open. An inverted cup 110, of greater diameter and length than the tube 108 is fitted around the tube 108, with an open end thereof reaching almost to the plate 66, while its closed end is sealed around the tube 98 at end 112. The tube, or inverted cup 76, also shown in Figure 4 and mentioned briefly with respect thereto, is of greater diameter and length than the tube or inverted cup 110, and fitted around this tube 110. The open end of the inverted cup or tube 76 is attached to the plate 66, while its closed end 124 is sealed about the tube 98. A set of nozzles 82, also shown in Figure 4, are attached about the exterior of the tube 76, near the top thereof. Each of these nozzles 82 communicates with the inside of the cup-like structure defined by the inverted cup of

tube 76, and has an exit directed downwardly towards the plate 66. The tube 76 is of larger diameter and length than the inverted cup 110, and fits around it. The lower end 116 of the inverted cup 110 reaches almost to the plate 66, where it communicates with the shift input. The top reaches above the catalyst bed, as indicated by reference numeral 88, where it communicates with the shift output.

In operation, effluent emerging from the tube 74 strikes the disc 96, causing radial outward flow of this effluent. The disc 96 may be rounded downwardly to minimize pressure loss, and to induce Coanda flow at its edges. Water entering the water entry tube 102 emerges through the hole 104 and also impacts the disc 96 on its upper surface, resulting in radial outward flow of the water. This radial outward flow may be aided by the porous spongy material which may optionally be located within the gap 106. The water and effluent meet at the edge of the disc 96 and, at this point, the high flow rate of the effluent entrains the water, with the resulting mixture flowing upward through annulus identified with reference numeral 118. Shear effects within the annulus 118 induce admixture and thermal equilibration. The mix then impacts the upper end 112 of the inverted cup 110, causing further mixing, and thereafter moving downwardly through annulus 120 between walls of the tube 108 and the inverted cup 110. Further shearing actions take place. The mix continues to move downward, until it impacts the metal plate 66, causing yet further mixing, and then moving into the annulus 122, located between the inverted cup 110 and inverted cup 76. Still further shear actions occur in this annulus 122. The mixture then leaves the inverted cup 76 through nozzles 82. Eduction of the gases from the exit (generally indicated as the space 124) of the shift reactor occurs as the mix moves into orifices 126, causing recirculation.

The dimensions of the shear mixer are set by the acceptable total pressure drop of the process, for example, 5 psi. These dimensions and pressure drops can be calculated through the use of normal engineering methods, and will not be discussed in further detail herein.

In experiments, it has been found that considerable amounts of solid carbon were produced when normal sprays were used to introduce water, while almost no solid carbon was formed using the shear mixing apparatus and methods as described above. The double nozzle assembly feature resulting in at least partial recirculation of the shift gases, proved amenable to engineering design and resulted in low pressure losses.

In the embodiment shown in Figure 5, the hydrated products pass from the shear mixing device into sets of nozzles 82, as described. These nozzles 82 are located within an open annulus 84 (defined by the tubes 86 and 76) that reaches all the way to the shift products at the top (see reference 124) and the water reservoir, indicated by water level 92, at the bottom of the heat exchanger. The nozzles 82 will educt shift material from the catalyst exit and recirculate it to the input, and this has the advantageous effect of adjusting the ratio of CO to CO₂. This can desirably effect product mix, and also increase the velocity of the shift gases which, in turn, can increase the heat transfer.

The effluent is then directed into larger orifices 126, increasing the degree of eduction provided by the nozzles 82. A ring 128 is attached to the outside of the inverted tube 76 just below the nozzles 82. The outer diameter of the ring 128 incorporates O-rings 130 that seal it to the interior of the tube 86. The orifices 52 are of a converging-diverging configuration, and there are an equal number of orifices 126 as there are nozzles 82. These orifices 126 are cut into the ring 128 at locations just below the nozzles 82. This modification brings the eduction process in line with conventional practice, which permits the dimensions of the nozzles 82 to be set by engineering parameters related to the amount of recirculation needed, as well as the parameters of the gas to be mixed.

With reference to Figure 6 of the drawings, it should be noted that hydrated process gases entering the shift reactor contain substantial non-reacted material, while those gases exiting the shift reactor contain very little non-reacted material. As a result (and based on both thermodynamics and

kinetics), the entering material is at a much higher level of reactivity than the material exiting. This suggests that increased shift efficiency would result from added catalyst content occurring at the end of the process. This added catalyst content can be accomplished by passing the process gases radially and outwardly through a cylindrical catalyst chamber, where the exiting gases contract a much greater volume of catalyst compared to normal vertical upward flow through a catalyst bed, of the type shown in Figure 4 of the drawings. Therefore, the embodiment shown in Figure 6 is a modification of that shown in Figure 4, and shows radially and outwardly moving gases, as opposed to vertically upward moving gases.

In Figure 6, the upward moving hydrated gases moving into the annulus designated with the reference numeral 132 enter into a distribution chamber 134. These gases then move radially outwardly towards the annulus 136 which is bounded at its inner surface by a porous cylinder 138, the porous cylinder defining the inner bounds of a catalyst chamber 140. The process gases then pass radially outwardly through the catalyst chamber bed 140, through a porous cylinder 142, and then finally into the annulus 136, which leads the shifted gases upward to space 144. This space 144 is defined on the bottom thereof by a plate 146, which also forms the top of the catalyst chamber 140. Thereafter, the shifted gases leave via an opening 148. The plate 146 can be lowered by force, or by gravity, to compensate for catalyst bed shrinkage, which normally occurs over an extended period of time.

The ratio of the volumes of catalyst encountered by the incoming process gases to that of the volume encountered by the exiting process gases is the square of the diameter ratios of the porous cylinders 142 and 138. In one embodiment, these may be 3 inches and 10 inches, resulting in a volume ration of 11. Such a ratio is sufficient to compensate for a decrease in shift activities over the shift process.

In a constant temperature heat exchanger, with shift reactor and fuel cell, a shift reactor with a fuel cell which both use constant temperature heat exchangers can be derived by

placing the fuel cell at the top of the apparatus as illustrated in Figure 4 of the drawings. In this way, the lower circular tube 18 is relocated from the top of the shift reactor to the fuel cell. The vertical tubes 24 are attached to the inlet of the water recirculation heat exchanger normally used to cool the fuel cell, and its output to the former inputs from vertical tubes 24 to the lower circular tube 18.

To operate this embodiment, air for operating the fuel cell enters the cell at its normal input, and the shifted hydrogen containing gases from the underoxidized burner enter via openings, as previously described. Heat from chemical reactions in the cell will be absorbed into the constant temperature heat exchanger, and dissipated to the environment via an external heat exchanger through the ends 52 and 54 of the control tube 50. Because of the intrinsic properties of the constant temperature heat exchanger, the fuel cell would remain at a very constant temperature throughout, and this results in improved cell efficiency.

Reference is now made to Figures 7 - 11 of the drawings, which show various embodiments and arrangements of the heat/mass exchanger of the invention.

In Figure 7, there is shown a heat/mass exchanger unit 160 that is fundamental to the humidified low temperature shift (HLTS) reactor, while Figure 8 has several of the heat/mass heat exchanger units 160 in a stacked condition, one on top of the other. The horizontal arrangement of these heat/mass heat exchangers 160 in stack form provides a heat/mass exchange assembly 162, shown in Figure 8. In Figure 7, water is introduced into water channel 164, which has a water inlet 166 and a water outlet 168. The water channel 164 is bounded at both its top and its bottom by a selectively porous layer, such as a hydrophobic layer 170, which is in contact with the shift catalyst 172. Water is introduced into the water channel 164 at the lowest suitable temperature for maximum hydrogen and minimum carbon monoxide production, through inlet 166, and flows in the direction of arrow A towards the water outlet 168.

A process gas channel 174 is provided on the side of the

hydrophobic layer 170 opposite to that of the water channel 164. The process gas channel 174 contains the catalyst 172. The process gas channel comprises an inlet 176 and an outlet 178, and the flow of the process gas is indicated by arrow B. Process gas thus passes from the inlet 176, into the process channel 174, passes through the catalytic material 172 and out through the outlet 178. The flow of process gas, as indicated by arrow B, is substantially countercurrent to the flow direction of the water, indicated by arrow A.

Water vapor produced in the water channel 166 passes through the hydrophobic layer 170. As previously mentioned, the hydrophobic layer is porous to vapors, but is impervious to liquid. Thus, liquid water flowing through the water channel 164 will not pass through the hydrophobic layer. However, water vapor produced by heating of the water within the water channel 164 passes through the hydrophobic membrane 170 to facilitate the shift reaction, resulting in the production of carbon dioxide, hydrogen and heat in the form of thermal energy. The thermal energy, indicated by arrow 180 moves through the hydrophobic layer 170 to provide energy, thereby vaporizing water flowing through the water channel 164. Water vapor moving from the water channel 164 to the process gas channel 174 is identified by reference numeral 182. The process continues, with the water moving towards the outlet 168, until it reaches the process gas inlet side of the shift.

In certain embodiments, there may be no need to provide a water outlet 168, since only water which is used in the system need be supplied.

With reference to Figure 8, this discloses essentially the same arrangement as that taught in Figure 7, except for the fact that the heat/mass exchanger units 160 are stacked to form a heat/mass exchange assembly 162, which is capable of processing larger volumes of process gas.

In Figure 9, there is shown an embodiment which, once more, is much the same disclosure as that in Figure 7. In this case however, the water channel is bounded only on one side thereof by the hydrophobic layer, and the process gas channel 174 has

both catalysts of normal activity, shown by reference numeral 184, and catalysts with higher activity, shown by reference numeral 186.

Figure 10 shows a detail of the shift reaction in the heat/mass exchanger. Carbon monoxide rich gas enters the process gas channel 174 and passes over the catalyst 172. The water vapor 182 flows from the liquid water channel 164 across the membrane 170, and reacts with the carbon monoxide to produce carbon dioxide, hydrogen and heat. The heat produced in this shift reaction flows back across the hydrophobic layer, indicated by reference numeral 180, and is used to heat the liquid water in the water channel 164, thereby providing more vapor for the shift reaction. Near the outlet 178 of the process gas channel, gas containing depleted carbon monoxide exits.

Reference is now made to Figure 8 of the drawings which illustrates a radial flow humidified low temperature shift (HLTS) reactor as an assembly of many sections 190. The multiple sections 190 produce a planar assembly 192. A gas stream containing hydrogen and carbon monoxide 194 enters the shift reactor 196 through an inlet pipe 198. The stream 194 thereafter enters the distribution channel 200 and thereafter the process gas inlets 202. A catalyst is located in each of the process gas channels, as shown in Figure 7 of the drawings. Gas flows through the catalyst radially outwardly towards the outlets 204.

On the other hand, water enters the HLTS reactor 196 through an inlet port 204 and flows down the water distribution channel 206. The water distribution channel 206 is connected to water inlets in each of the sections 190, in much the same way as shown with respect to the connection of each water inlet in Figure 7 of the drawings. Appropriate liquid seal rings 206 are provided in the catalyst layer, and holes through the hydrophobic layer 208 are provided.

Water from the channel 204 enters inlets and flows countercurrent and radially inward towards water outlets 210. An appropriately sealed distribution gas manifold 212, with appropriate seals 214, collects the gases from the units, once they have passed through the outlets. Water exits through a

cavity 216, and enters a liquid collection manifold 218. Water from the manifold 218 exits the reactor through the outlet port 220.

5 Multiple liquid flow distribution channels may be used to enhance flow uniformity. The process gas exits the catalyst layer and passes out through the outlets, entering the distribution manifold 212, flowing out of the reactor through a pipe 222. The distribution manifold 212 is formed as a space between the outside edges of the planar structures which
10 comprises the heat mass exchanger assemblies on the one hand, and a housing 224. A plate 226 is provided which applies a compressive downward force on the various assemblies mounted on the support plate 228 to ensure flow uniformity and proper sealing of the liquid flow and gas distribution manifold.

15

The invention is not limited to the precise details herein described, and modifications thereof may be made within the scope of the claims.

CLAIMS:

1. A heat exchanger comprising:
 - a housing defining a chamber;
 - a holding system within the chamber, the holding system
 - 5 comprising sealed passages for accommodating a liquid vapor medium,
 - a liquid-vapor circulating system located within the holding system, the circulating system having conduits extending through the passages of the holding system, the conduits for
 - 10 accommodating a liquid-vapor medium;
 - circulating means for circulating liquid-vapor medium in the conduits of the circulating system;
 - wherein heat is exchanged between the chamber of the housing and the holding system, and the housing and the
 - 15 circulating system, with the circulating system spreading the heat throughout the housing to provide a substantially constant temperature therethrough.
2. A heat exchanger as claimed in claim 1 wherein the holding system comprises:
 - 20 an upper circular tube defining a passage;
 - a lower circular tube defining a passage;
 - a vertical tube having a passage, the vertical tube being connected at ends thereof to the upper and lower circular tubes respectively such that the passages of the vertical tube and
 - 25 upper and lower circular tubes are in communication with each other.
3. A heat exchanger as claimed in claim 1 wherein the circulating system comprises an inner circular tube located in a passage of at least one of the upper or lower circular tubes.
- 30 4. A heat exchanger as claimed in claim 3 wherein the circulating system comprises an inner circular tube located within the passage of both of the upper and lower circular tubes respectively.
5. A heat exchanger as claimed in claim 4 further
- 35 comprising at least one side arm tube extending between the inner circular tube in the upper circular tube and the inner circular tube in the lower circular tube.

6. A heat exchanger as claimed in claim 5 further comprising a pump means in the side arm tube, the pump means facilitating circulation of a water-vapor medium in passages of the side arm tube and inner circular tubes respectively.

5 7. A heat exchanger as claimed in claim 1 further comprising a second liquid-vapor circulating system in at least part of the holding system, the second liquid vapor circulating system accommodating a liquid-vapor medium and having ducts for connecting to an external source for regulating the temperature
10 of liquid-vapor medium within the second circulating system.

8. A heat exchanger as claimed in claim 7 wherein the second liquid-vapor circulating system comprises a circular tube located within the passage of the upper circular tube, the ducts for connecting to an external source extending outwardly from
15 the upper circular tube.

9. A shift reactor and heat exchanger system comprising:
a housing defining a chamber which contains a catalyst, the catalyst decreasing carbon monoxide and /or increasing hydrogen production in a product gas passing therethrough,
20 a holding system within the chamber, the holding system comprising sealed passages for accommodating a liquid-vapor medium,

a liquid-vapor circulating system located within the holding system, the circulating system having conduits extending
25 through the passages of the holding system, the conduits for accommodating a liquid-vapor medium;

circulating means for circulating liquid-vapor medium in the conduits of the circulating system;

wherein heat is exchanged between the inside of the housing
30 and the holding system, and the housing system and the circulating system, with the circulating system spreading the heat throughout the housing to provide a substantially constant temperature therethrough.

10. A shift reactor and heat exchanger system as claimed
35 in claim 9 further comprising shear mixing and recirculating means in the chamber through which product gas enters the chamber and is thoroughly mixed with water before reaching the

catalyst in the chamber.

11. A shift reactor and heat exchanger system as claimed in claim 10 wherein the shear mixing and recirculating means comprises a series of concentric tubes axially mounted with respect to each other, the concentric tubes providing a continuous pathway defined by an annulus between each pair of adjacent tubes, the pathway forcing a gas mixture therein into reverse flows.

12. A shift reactor and heat exchanger system as claimed in claim 10 wherein the shear mixing and recirculating means further comprises:

a disc member, the disc member being located in a path of effluent introduced into the shear mixing and recirculating means to cause it to flow radially outward, and

a water entry means for introducing water onto an opposite side of the disc, the water flowing radially outwardly therefrom to mix with the gas stream.

13. A shift reactor and heat exchanger system as claimed in claim 12 wherein the shear mixing and recirculating means comprises at least one nozzle through which the process gas/water mixture exits from the shear mixing and recirculating means, the nozzles being located so as to educt and recirculate gases through the shift reactor and heat exchanger system.

14. A shift reactor and heat exchanger system as claimed in claim 9 wherein an upper screen is located in the chamber at the upper level of the catalyst and a lower screen is located in the chamber defining the lower level of the catalyst.

15. A shift reactor and heat exchanger system as claimed in claim 9 further comprising a reservoir at the lower end of the chamber for receiving and holding water through which the product gas passes before reaching the catalyst.

16. A shift reactor and heat exchanger system as claimed in claim 9 wherein the system is structured so as to cause the product gases to flow substantially vertically and upwardly through the catalyst in the chamber.

17. A shift reactor and heat exchanger system as claimed in claim 9 wherein the system is structured so as to cause the

product gases to flow substantially radially and outwardly through the catalyst in the chamber.

18. A shift reactor and heat exchanger system as claimed in claim 17 wherein the chamber comprises an inner porous cylinder and an outer porous cylinder defining a space within which the catalyst is located, means for introducing the product gas through the inner porous system, and an annulus spaced radially outwardly of the outer porous cylinder through which the treated product gas is collected.

10 19. A heat/mass exchanger comprising:

a water conduit having an inlet and an outlet so that water flows through the water conduit in a first direction;

a layer at least partially surrounding the water conduit, the layer being substantially impervious to liquid water and permitting free flow of water vapor therethrough;

15 a process gas conduit containing a catalyst for decreasing carbon monoxide and/or increasing hydrogen production in a process gas stream passing therethrough, the process gas conduit at least partially surrounding the layer and having an inlet and an outlet defining a flow path for the process gas stream in a second direction.

20 20. A heat/mass exchanger as claimed in claim 19 wherein the inlet and outlet of the water conduit, and the inlet and outlet of the process gas conduit are arranged such that the water flow in the first direction is substantially countercurrent to the process gas stream flow in the second direction.

21. A heat/mass exchanger as claimed in claim 20 wherein the layer between the water conduit and the process gas conduit is comprised of a hydrophobic layer.

22. A heat/mass exchanger stack comprising a plurality of mass exchangers as claimed in claim 20.

23. A heat/mass exchanger as claimed in claim 19 wherein the process gas conduit comprises a first catalyst having normal activity, and a second catalyst of higher activity.

24. A heat exchanger and shift reactor system comprising: a plurality of heat exchange members each having a water

conduit with an inlet and an outlet so that water flows through the water conduit in a first direction, a layer at least partially surrounding the water conduit, the layer being substantially impervious to liquid water and permitting substantially free flow of water vapor therethrough, a process gas conduit containing a catalyst for decreasing carbon monoxide and/or increasing hydrogen production in a process gas stream passing therethrough, the process gas conduit at least partially surrounding the layer and having an inlet and an outlet defining a flow path in a second direction;

a water inlet manifold connectable to a source of water and for supplying water to the inlet of each water conduit;

an inlet distribution channel for receiving a process gas stream from a source and connected to each process gas conduit;

an outlet distribution channel connected to the outlet of each process gas conduit.

25. A heat exchanger and shift reactor system as claimed in claim 24 wherein the layer comprises a hydrophobic layer.

26. A heat exchanger and shift reactor system as claimed in claim 24 wherein the water flow in the first direction is substantially countercurrent to the process gas flow in the second direction.

27. A heat exchanger and shift reactor system as claimed in claim 24 further comprising a water collection manifold connected to the outlet of each water conduit.

28. A heat exchanger and shift reactor system as claimed in claim 24 further comprising liquid seal rings.

29. A heat exchanger and shift reactor system as claimed in claim 24 further comprising a housing containing the heat exchange members, a water inlet manifold, and inlet and outlet distribution channels for the process gas, the housing further comprising an upper plate which provides a downward force on the exchange members to ensure flow uniformity and proper sealing catalyst and flow.

30. A process for controlling temperature distribution in a heat exchanger having a housing defining a chamber, the process comprising:

forming a framework of passages in the chamber and filling the passages with a water-vapor medium such that heat transfer due to thermal changes can occur between the chamber and the water-vapor medium in the passages;

5 forming a framework of conduits in at least a portion of the passages and filling the conduits with a water-vapor medium such that heat transfer due to thermal changes can occur between the water-vapor medium in the passages and the water-vapor medium in the conduits;

10 circulating the water-vapor medium in the conduits whereby thermal changes in the chamber are distributed substantially evenly through the chamber by the water-vapor mediums in the passages and conduits respectively to provide a substantially controlled and constant temperature in the chamber.

15 31. A process as claimed in claim 30 wherein the framework of passages is formed by circular and linear tubes so that the passages extend to substantially all areas within the housing.

20 32. A process for treating a gas mixture containing carbon monoxide by raising the humidity level and lowering the temperature of the gas mixture, the process comprising the steps of:

25 passing the gas mixture in a first direction through a gas mixture conduit containing a catalyst, the catalyst being selected for its ability to decrease carbon monoxide and/or increase hydrogen production in the gas mixture passing therethrough;

 passing water in a second direction through a water conduit adjacent the gas mixture conduit,

30 inserting a layer between the water conduit and gas mixture conduit, the layer being impervious to liquid water but permitting free flow of water vapor thereacross;

 transferring thermal energy of the gas mixture to the water causing evaporation of at least a part of the water to water vapor and reducing the temperature of the gas mixture;

35 transferring water vapor to the gas mixture to thereby maintain and/or increase water vapor partial pressure of the gas mixture;

wherein water vapor passes from the water conduit to the gas mixture conduit to facilitate a shift reaction converting at least a portion of the carbon monoxide into hydrogen, carbon dioxide and heat, the heat vaporizing the water in the water conduit to reduce temperature of the gas mixture and provide water vapor to the gas mixture for further shift reactions.

33. A process as claimed in claim 32 wherein the second direction of the water is substantially countercurrent to the first direction of the gas mixture.

Equilibrium CO Concentration (Gasoline/air UOB Reformer; 0.37 Stoichiometric Ratio; 16 psia Pressure)

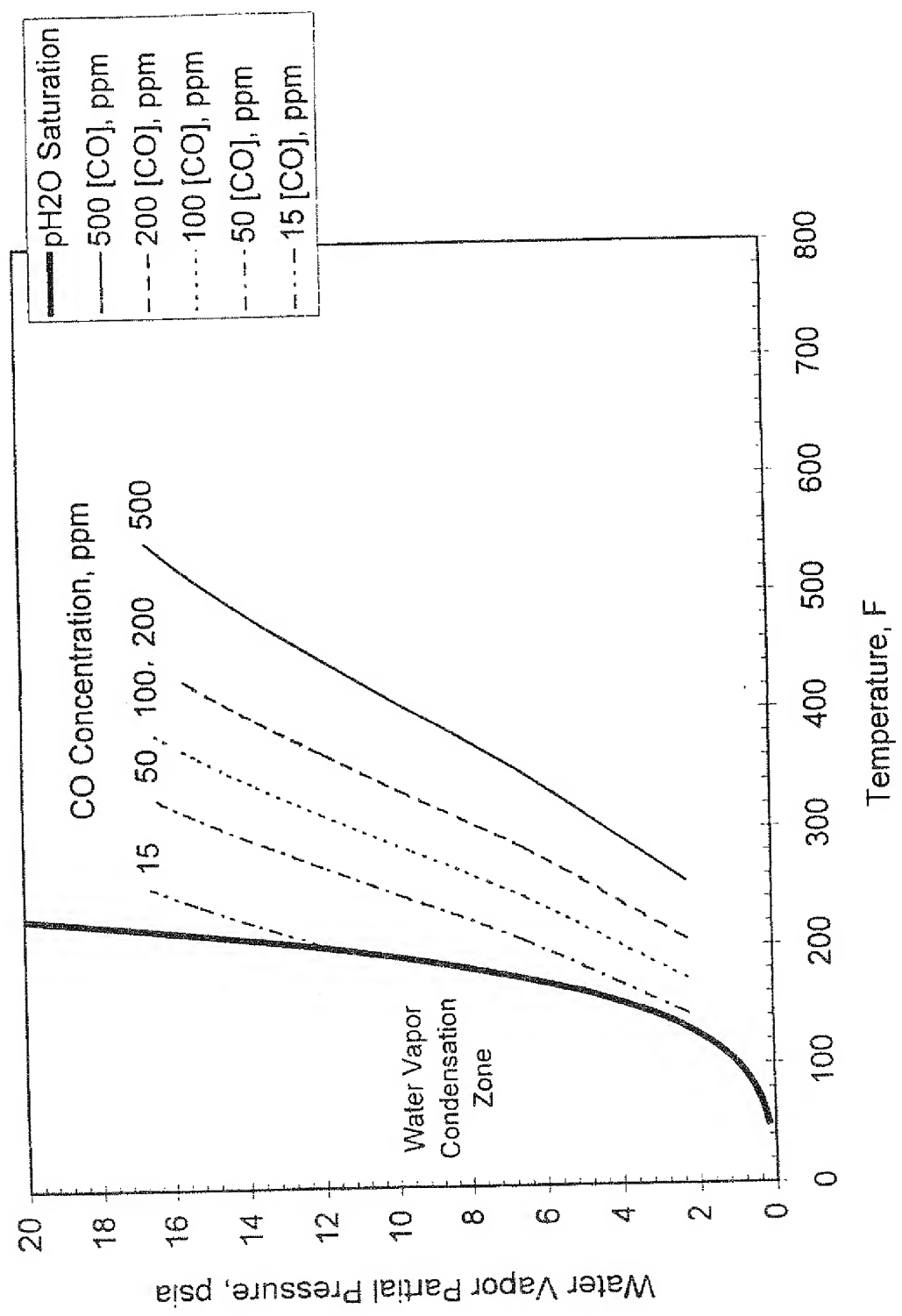


FIG. 1

Comparison of Various Low Temperature Shift (LTS) Processes
(Gasoline/air UOB Reformer; 0.37 Stoichiometric Ratio; 16 psia Pressure)

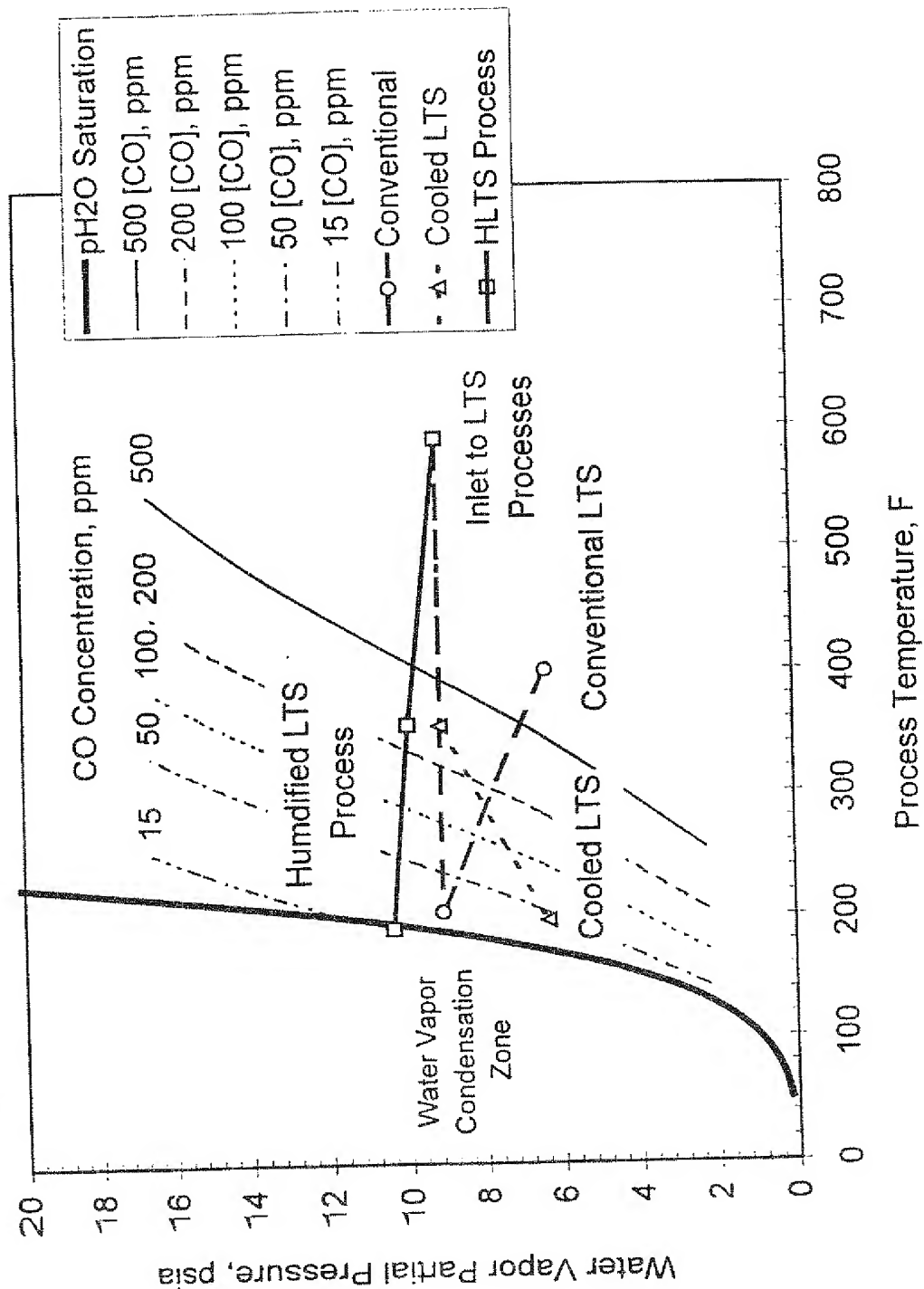


FIG. 2

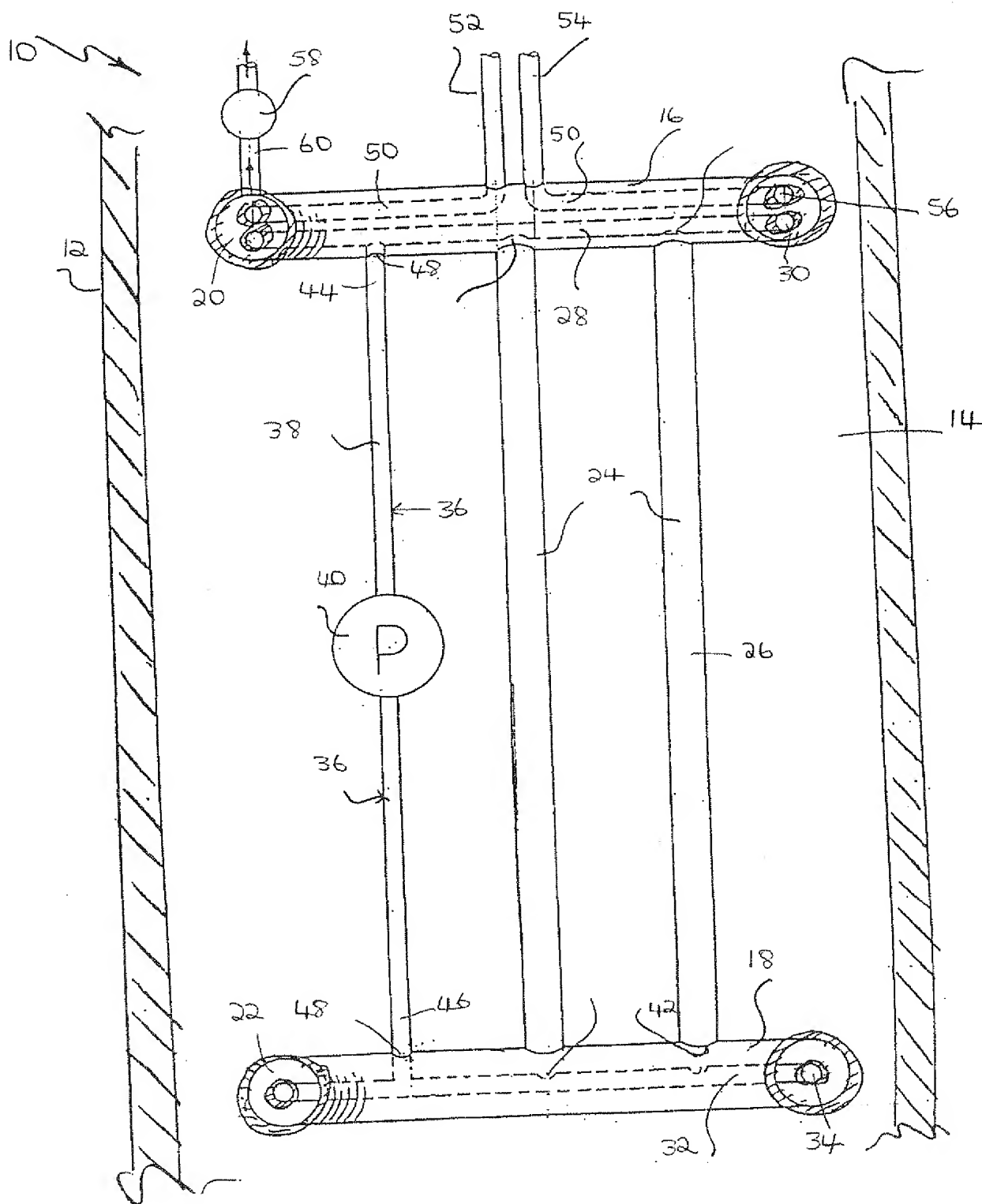


FIG. 3

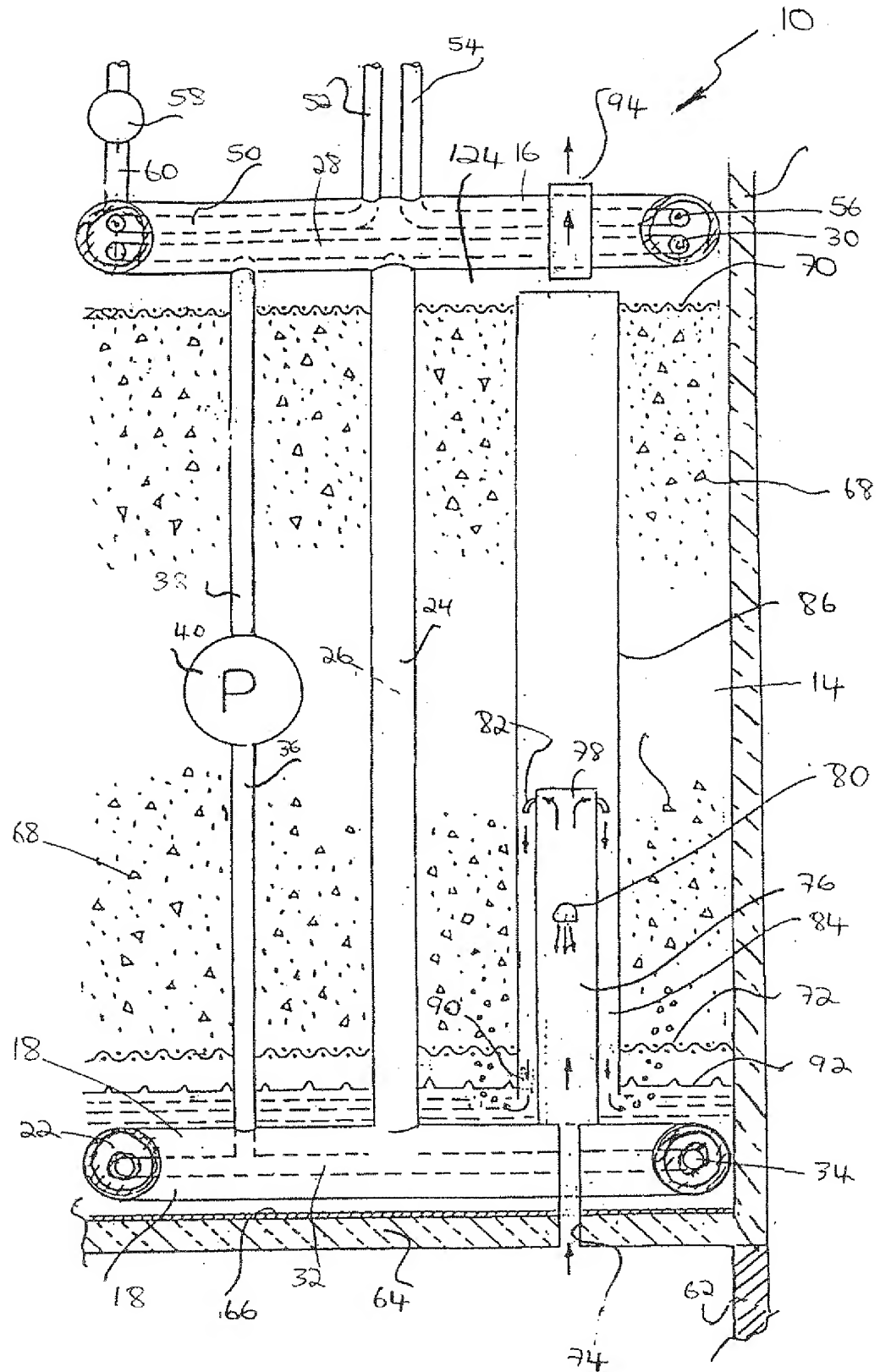


FIG. 4

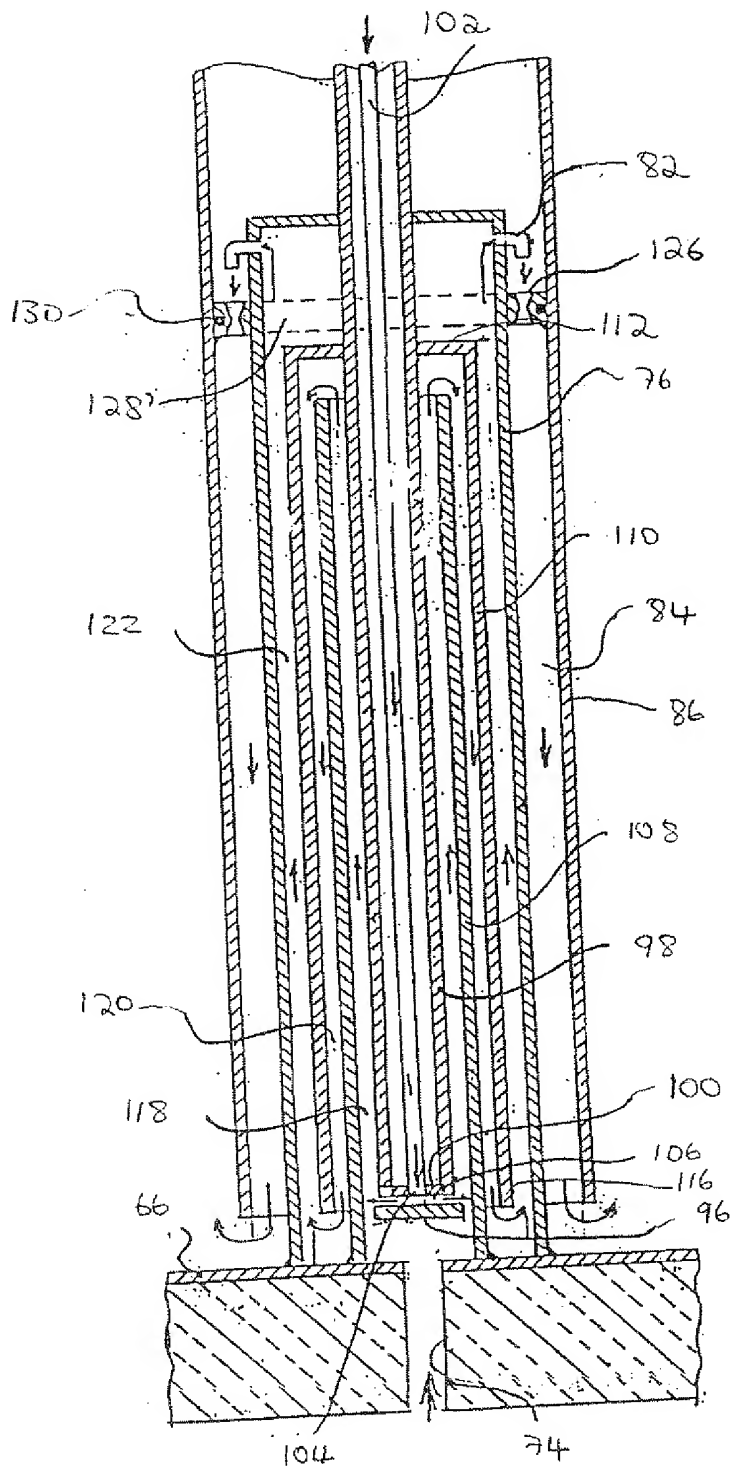


FIG. 5

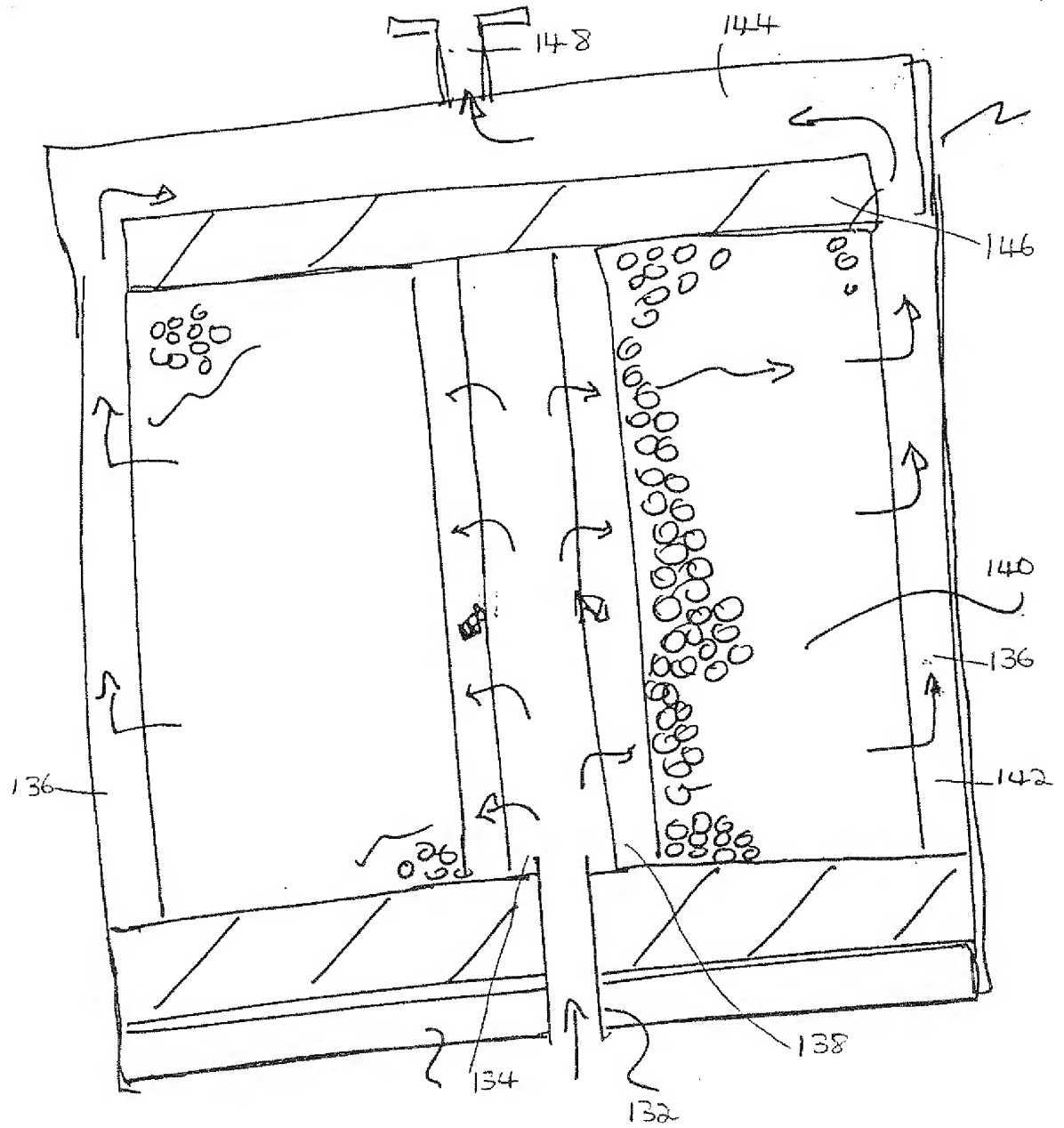


FIG. 6

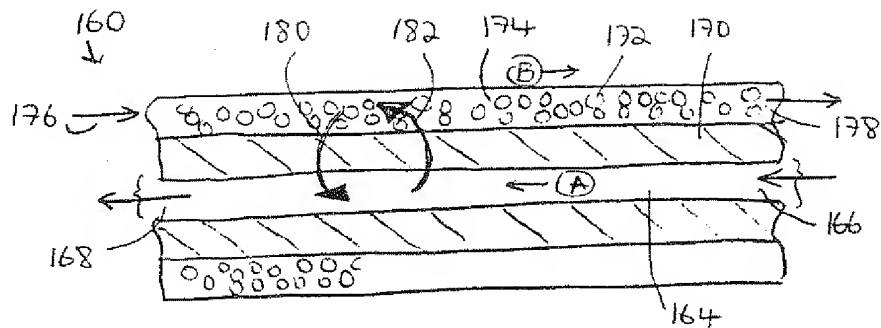


FIG. 7

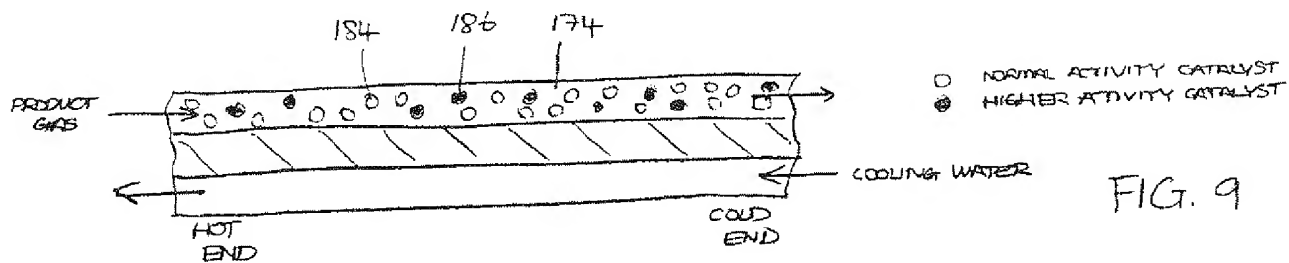


FIG. 9

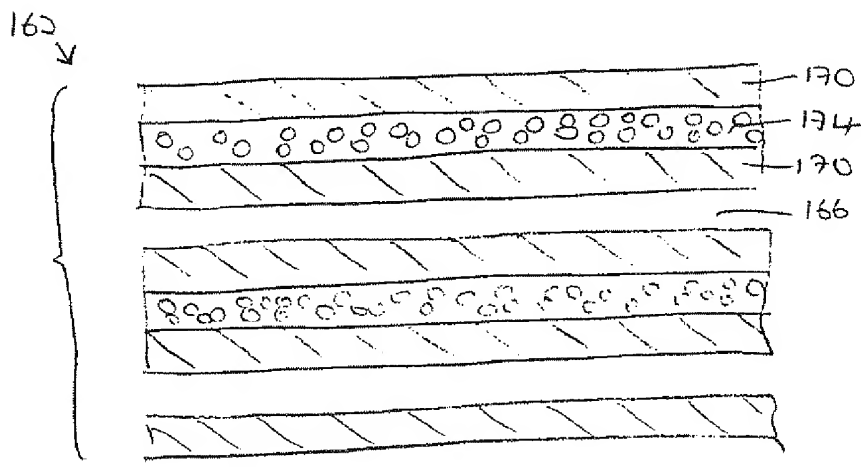


FIG. 8

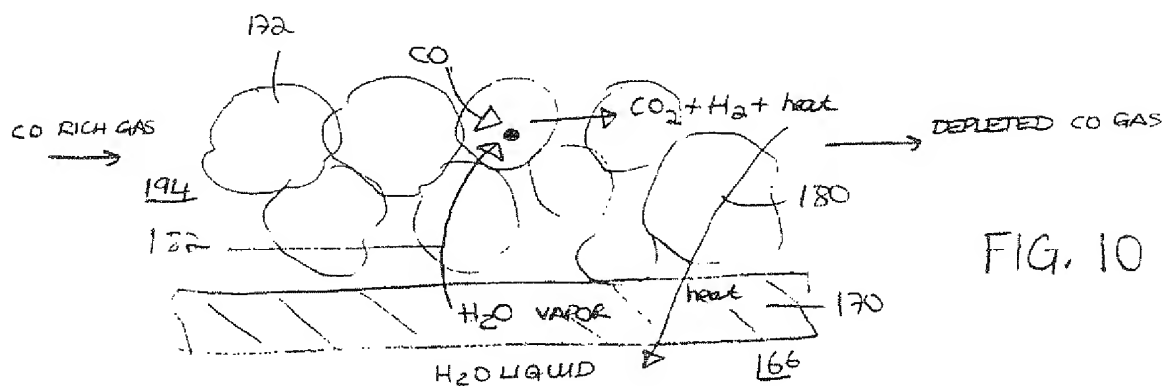


FIG. 10

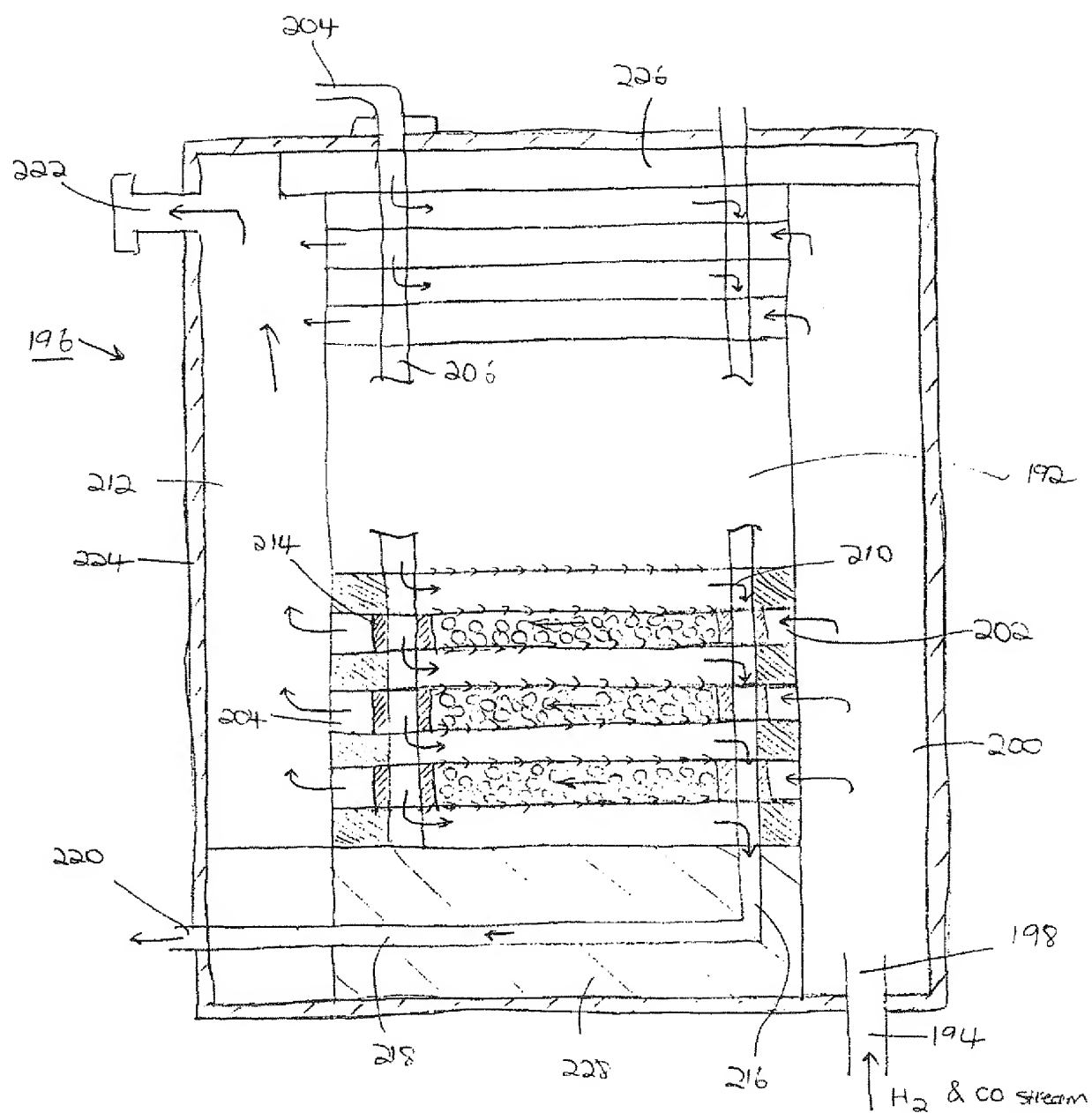


FIG. 11